

AMENDMENT UNDER 37 C.F.R. § 1.111
Application Serial No. 09/582,495

REMARKS

Claims 1-14 and 19-20 are all the claims pending in the application. Support for the amendments to the claims in the instant response can be found in the specification on page 8, line 14 to page 9, line 2.

I. Status of the Claims

Applicants note that the Office Action Summary sheet indicates that claims 1-14 are pending in the application, but that claims 1-14 have been rejected and claims 19-20 are allowed. However in the body of the Office Action, claims 1-14 were rejected and claims 19-20 are indicated as allowed. Applicants believe that claims 1-14 and 19-20 are the claims pending in the Office Action and request clarification. For purposes of this Amendment, claims 1-14 are viewed as rejected and claims 19-20 are viewed as objected to as dependent upon rejected base claims.

II. Priority

Applicants thank the Examiner for acknowledging Applicants' claim for foreign priority under 35 U.S.C. § 119 (a)-(d) and Applicants' claim for domestic priority under 35 U.S.C. § 119(e). However, the Examiner did not indicate whether the priority documents had been received. We note that this is a National Stage Application filed under 35 U.S.C. § 371 and the Notification of Acceptance of Application under 35 U.S.C. § 371 dated August 15, 2000, also does not indicate that the priority documents have been received. Since the International Bureau should have sent the certified copies to the PTO, Applicants respectfully request formal acknowledgment of receipt of the priority documents.

AMENDMENT UNDER 37 C.F.R. § 1.111
Application Serial No. 09/582,495

III. Claim Rejections Under 35 U.S.C. § 112

Claims 1 and 5 were rejected under 35 U.S.C. 112, second paragraph, as being indefinite. Specifically the Examiner states that in formula (1) of claim 1, the definition of R is not included and in the definition of the groups of formula (1), R⁵ is included but not indicated in the general structure.

Regarding claim 5, the Examiner states that the recitation of a process "wherein the concentration of the unsaturated group-containing ester represented by general formula (1) at the time of the hydrogenation reaction thereof is on the range of 1 wt % -50 wt % based on the entirety of the raw material liquid containing the unsaturated group-containing ester" is unclear because the phrase "at the time of the hydrogenation reaction" could be at the initial time of the reaction or any other time.

In regard to claim 1, Applicants submit that there was a typographical error in the structure of formula (1) such that the fifth R is not identified. Because the discrepancy occurs in the structure, claim 1 is cancelled herein and new claim 24, directed to the subject matter of claim 1 with the proper structure indicating the fifth R as R⁵ is added. Support can be found in the specification on page 8. Applicants submit that one of ordinary skill in the art would recognize the typographical error since the R¹, R², R³, R⁴, and R⁶ are each identified and the claim recites a definition for an R⁵ group and because the structure is identified in the disclosure. This change is not made for purposes of patentability.

Claim 5 has been cancelled herein and therefore the rejection is rendered moot. However, as applied to the new and amended claims, Applicants have more clearly specified that

AMENDMENT UNDER 37 C.F.R. § 1.111
Application Serial No. 09/582,495

that concentration of the unsaturated group-containing ester represented by formula (1) is at the initial time of the reaction. Therefore, withdrawal of the rejection is respectfully requested.

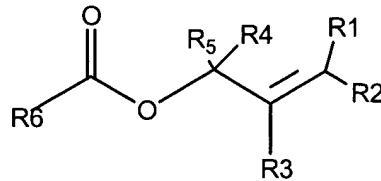
IV. Claim Rejections under 35 U.S.C. § 102

A. Aldrich Catalog

Claim 14 was rejected under 35 U.S.C. 102(b) as being anticipated by Aldrich Catalog, Handbook of Fine Chemicals, 1996-97. According to the Examiner, Aldrich teaches the commercial availability of a variety of esters, including propyl acetate, propyl propionate, butyl acetate, butyl propionate, ethyl acetate, ethyl propionate (Aldrich Catalog, pages 263, 280, 667, 707, 1269, 1273).

B. Tanaka, et al.

Claims 1, 2, 5, 6, 7, 8, 9, 10, 11, 12, 13 and 14 were rejected under 35 U.S.C. 102(b) as being anticipated by Tanaka et al JP09-194427. The Examiner states that Tanaka et al teach a method for the production of saturated esters by hydrogenation of unsaturated esters having the general formula (page 2):



wherein R1 to R5 represent a hydrogen atom or any alkyl group having 1 to 10 carbon atoms which may be the same or different and R6 is a straight-chain or branched alkyl group having 1 to 10 carbon atoms (page 3).

The Examiner further states that Tanaka et al teach a process comprising:

AMENDMENT UNDER 37 C.F.R. § 1.111
Application Serial No. 09/582,495

1. a hydrogenation reaction that can be carried out in liquid phase or gas phase, using a variety of reactors such as complete mixed tank, bubble tower, trickle bed and the like (page 8);
2. the use of a variety of unsaturated esters in the hydrogenation reaction, including allyl acetate, crotyl acetate, methallyl acetate, allyl propionate, crotyl propionate and methallyl propionate (page 3);
3. the use of hydrogenation catalysts such as rhodium catalysts and that nickel and other metal catalyst such as Cu, Cr, Mg, and Al may be used because of the fact that rhodium is expensive (pages 5,7,8);
4. a reaction temperature range from 30 to 200 °C (page 8,9); and
5. a method for the production of saturated esters like propyl acetate, butyl acetate, propyl propionate, and butyl propionate among others (page 9).

C. Jiang et al.

Claims 1 to 4 were rejected under 35 U.S.C. 102(b) as being anticipated by Jiang et al, Journal of Shanghai University of Science and Technology, no. 1, pages 81-87, 1987. The Examiner asserts that Jiang et al teach:

1. the hydrogenation of different unsaturated compounds using a catalyst comprising palladium (page 82); and
2. the catalytic hydrogenation of vinyl acetate in ethyl acetate as a solvent (see table 3, page 84).

AMENDMENT UNDER 37 C.F.R. § 1.111
Application Serial No. 09/582,495

With respect to the rejection over the Aldrich Catalogue, claim 14 is cancelled herein and therefore the rejection as to claim 14 is moot.

With respect to the rejection over Tanaka et al and Jiang et al, Applicants respectfully traverse the Examiner's rejections and submit that none of the cited references teach or suggest the presently claimed invention for the following reasons.

The present invention relates to a process for producing a hydrogenated ester by hydrogenating an unsaturated group-containing ester of a specific formula (1) in the presence of a hydrogenating catalyst so as to produce a corresponding hydrogenated ester, comprising:

providing an unsaturated group-containing ester (1) wherein the concentration of the unsaturated group-containing ester of formula (1) at the initial time of the hydrogenation reaction thereof is in the range of 1 wt.% - 50 wt.% based on the entirety of the raw material liquid; and reacting the unsaturated group-containing ester by diluting said unsaturated group-containing ester with an inert solvent.

As described above, in the process for producing a hydrogenated ester according to the present invention, a hydrogenated ester (particularly, a saturated ester) can be industrially produced while maintaining the resultant raw material conversion, selectivity factor, and yield to a high level. In the present invention, a complicated reaction apparatus or reaction process is not necessarily required.

Further, according to the present invention, saturated esters can industrially be produced at a low cost by conducting a hydrogenation reaction in a liquid phase by use of a hydrogenating

AMENDMENT UNDER 37 C.F.R. § 1.111
Application Serial No. 09/582,495

catalyst commonly used, and by use of a raw material liquid which has been obtained by diluting an unsaturated group-containing ester with a solvent inert to the hydrogenation reaction (page 75, line 27 to page 76, line 4 of the present specification).

None of the cited references teach or suggest a process wherein an unsaturated group-containing ester of formula (1) is hydrogenated at a concentration of the unsaturated group-containing ester at the initial time of the hydrogenation reaction in the range of 1 wt.% - 50 wt.%, while the unsaturated group-containing ester is diluted with an inert solvent.

In this regard JP09-194427 (Daicel Chemical discloses a production process of saturated esters, which is characterized by hydrogenation of a specific unsaturated group-containing ester with a hydrogenation catalyst containing 10 -100 wt.% of nickel (claim 1 of JPO9-194427).

However, this reference does not teach or suggest a process wherein an unsaturated group-containing ester of formula (1) is hydrogenated at a concentration of the unsaturated group-containing ester at the initial time of the hydrogenation reaction in the range of 1 wt.% - 50 wt.%, while the unsaturated group-containing ester is diluted with an inert solvent.

More specifically, according to the present inventors' experiments, when the method of this reference (JPO9-194427) is used, it is difficult to conveniently obtain a saturated ester having a high purity. In other words, in the method of this reference, the selectivity factors for the saturated ester and the carboxylic acid cannot be made 98.0% or more and 2.0% or less, respectively, when the conversion of the unsaturated group-containing ester is made 99.8% or more (as stated on page 6, line 19 to page 7, line 22 of the present specification).

AMENDMENT UNDER 37 C.F.R. § 1.111
Application Serial No. 09/582,495

On the contrary, according to the present invention, the selectivity factors for the saturated ester and the carboxylic acid are made 98.0% or more and 2.0% or less, respectively, when the conversion of the unsaturated group-containing ester is made 99.8% or more (Table 1, on page 46 of the present specification).

In regard to Jiang (Journal of Shanghai University of Science), silica-supported polyvinyl-p-dimethylamino benzal-metal complexes comprising metal atom Pt or Pd which have catalytic activity for hydrogenation of unsaturated acids are disclosed. (Abstract). However, this reference does not teach or suggest a process wherein an unsaturated group-containing ester of formula (1) is hydrogenated at a concentration of the unsaturated group-containing ester at the initial time of the hydrogenation reaction in the range of 1 wt.% - 50 wt.%, while the unsaturated group-containing ester is diluted with an inert solvent.

Specifically, independent claim 10 relates to a process for producing a hydrogenated ester by hydrogenating an allyl-type ester of formula (1) ($n = 1$) by using a hydrogenating catalyst so as to produce a corresponding allyl-type ester, wherein the concentration of a carboxylic acid in a raw material containing the allyl-type ester of formula (1) is 1 wt.% or less.

JP09-194427 (Daicel Chemical) discloses a production process of saturated esters as described above. However, this reference does not teach or suggest a process using an allyl-type ester wherein the concentration of a carboxylic acid in the raw material is 1 wt.% or less.

Thus, the cited references do not teach or suggest the claimed invention. Accordingly Applicants respectfully request withdrawal of the rejections.

AMENDMENT UNDER 37 C.F.R. § 1.111
Application Serial No. 09/582,495

IV. Allowable Subject Matter

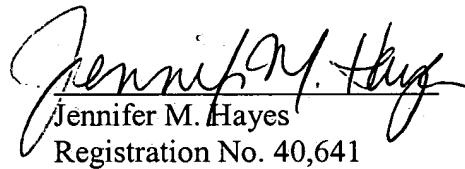
On page 5 of the Office Action, the Examiner indicates claims 19 and 20 as allowable if rewritten in independent format to include the limitations of the base claims and any intervening claims. Claims 19 and 20 are cancelled herein and in view thereof, Applicants have added new claims 31-32 directed to the subject matter of claim 19. New claims 33-34 are also added directed to the subject matter of claim 20. Accordingly, Applicants respectfully request withdrawal of the objection.

V. Conclusion

In view of the above, reconsideration and allowance of this application are now believed to be in order, and such actions are hereby solicited. If any points remain in issue which the Examiner feels may be best resolved through a personal or telephone interview, the Examiner is kindly requested to contact the undersigned at the telephone number listed below.

Applicant hereby petitions for any extension of time which may be required to maintain the pendency of this case, and any required fee, except for the Issue Fee, for such extension is to be charged to Deposit Account No. 19-4880.

Respectfully submitted,


Jennifer M. Hayes
Registration No. 40,641

SUGHRUE, MION, ZINN,
MACPEAK & SEAS, PLLC
2100 Pennsylvania Avenue, N.W.
Washington, D.C. 20037-3213
Telephone: (202) 293-7060
Facsimile: (202) 293-7860
Date: October 5, 2001

APPENDIX
VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE CLAIMS:

Claims 1,2, 5, 14, and 19-20 are canceled.

The claims are amended as follows:

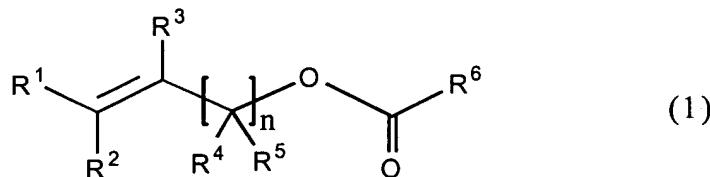
3. (Amended) A process for producing a hydrogenated ester according to claim 2 30, wherein the inert solvent is the corresponding hydrogenated ester.
6. (Amended) A process for producing a hydrogenated ester according to any of claims 2-5 claim 30, wherein the reaction temperature at the initial time of the hydrogenation reaction is in the range of 0°C-200°C.
7. (Amended) A process for producing a hydrogenated ester according to any of claims 2-6 claim 30, wherein the unsaturated group-containing ester represented by the general formula (1) is at least one compound selected from the group consisting of: allyl acetate, crotyl acetate, methallyl acetate, allyl propionate, crotyl propionate, methallyl propionate, vinyl acetate, vinyl propionate, 1,3-butadienyl acetate, and 1,3-butadienyl propionate.
8. (Amended) A process for producing a hydrogenated ester according to any of claims 2-7 claim 30, wherein the hydrogenating catalyst comprises at least one element selected from the group consisting of Group VIII elements, Group IX elements or and Group X elements in the periodic table.
9. (Amended) A process for producing a hydrogenated ester according to any of claims 2-8 claim 30, wherein the hydrogenation reaction, is conducted by a liquid-phase reaction by use of a fixed bed-type reactor.

AMENDMENT UNDER 37 C.F.R. § 1.111
Application Serial No. 09/582,495

13. (Amended) A process for producing a hydrogenated ester according to ~~any of claims 10-12~~ claim 10, wherein the allyl-type ester represented by the general formula (1) is at least one species of allyl-type ester selected from the group consisting of allyl acetate, crotyl acetate, methallyl acetate, allyl propionate, crotyl propionate, or and methallyl propionate.

Claims 30-34 are added as new claims.

30. (New) A process for producing a hydrogenated ester by hydrogenating an unsaturated group-containing ester represented by the following general formula (1) in the presence of a hydrogenating catalyst so as to produce a hydrogenated ester corresponding to the unsaturated group-containing ester:



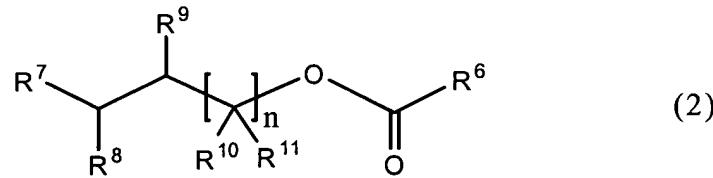
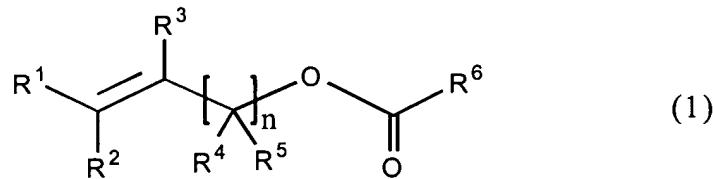
wherein R¹, R², R³, R⁴ and R⁵ denote an arbitrary alkyl group containing 1-10 carbon atoms, an arbitrary alkenyl group containing 1 - 10 carbon atoms, or a hydrogen atom and may be the same as or different from each other; the alkyl group and alkenyl group may be either straight-chain or branched; R⁶ denotes an arbitrary alkyl group which contains 1 - 10 carbon atoms and may be either straight-chain or branched; and n is 0 or 1, comprising

providing an unsaturated group-containing ester represented by the general formula (1), wherein the concentration of the unsaturated group-containing ester represented by general formula (1) at the initial time of the hydrogenation reaction thereof is in the range of 1 wt % -50 wt % based on the entirety of the raw material liquid containing the unsaturated group-containing ester; and

AMENDMENT UNDER 37 C.F.R. § 1.111
Application Serial No. 09/582,495

reacting the unsaturated group containing ester by diluting said unsaturated group-containing ester with an inert solvent to effectuate a hydrogenation reaction.

31. (New) A process for producing a hydrogenated ester, wherein an unsaturated group-containing ester represented by the general formula (1) is hydrogenated by using a hydrogenating catalyst which contains at least one metal selected from the group consisting of Group VIII elements, Group IX elements, and Group X elements in the periodic table, and is to be used for hydrogenating an unsaturated group-containing ester represented by the following formula (1) to thereby produce a hydrogenated ester represented by the following formula (2), wherein the catalyst has an acidity of 1.0×10^{-1} mol/g or less:



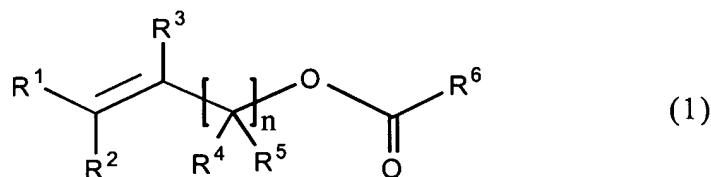
wherein n represents 0 or 1; R¹, R², R³, R⁴ and R⁵ denote an arbitrary alkyl group containing 1-10 carbon atoms, an arbitrary alkenyl group containing 1 - 10 carbon atoms, or a hydrogen atom and may be the same as or different from each other; the alkyl group and alkenyl group may be either straight-chain or branched; R⁶ represents a C₁-C₁₀ alkyl group; and each of R⁷, R⁸, R⁹, R¹⁰,

AMENDMENT UNDER 37 C.F.R. § 1.111
Application Serial No. 09/582,495

and R¹¹ represents a C₁-C₁₀ alkyl group, a C₁-C₁₀ alkenyl group, or a hydrogen atom independently to each other.

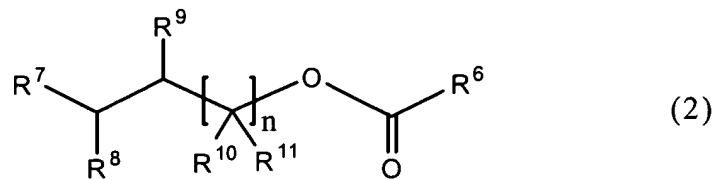
32. (New) A process for producing a hydrogenated ester according to claim 31, wherein the hydrogenating catalyst selected from at least one metal selected from the group consisting of Group VIII elements, Group IX elements, and Group X elements in the periodic table is at least one species selected from the group consisting of palladium, ruthenium and rhodium.

33. (New) A process for producing a hydrogenated ester, wherein at least one species of an unsaturated group-containing ester selected from the group consisting of allyl acetate, crotyl acetate, methallyl acetate, allyl propionate, crotyl propionate, methallyl propionate, vinyl acetate, 1,3-butadienyl acetate, 1-methyl-1-propenyl acetate, vinyl propionate, 1,3-butadienyl propionate, and 1-methyl-1-propenyl propionate is hydrogenated by using a hydrogenating catalyst which contains at least one metal selected from the group consisting of Group VIII elements, Group IX elements, and Group X elements in the periodic table, and is to be used for hydrogenating an unsaturated group-containing ester represented by the following formula (1) to thereby produce a hydrogenated ester represented by the following formula (2), wherein the catalyst has an acidity of 1.0 x 10⁻¹ mol/g or less:



AMENDMENT UNDER 37 C.F.R. § 1.111

Application Serial No. 09/582,495



wherein n represents 0 or 1; R¹, R², R³, R⁴ and R⁵ denote an arbitrary alkyl group containing 1-10 carbon atoms, an arbitrary alkenyl group containing 1 - 10 carbon atoms, or a hydrogen atom and may be the same as or different from each other; the alkyl group and alkenyl group may be either straight-chain or branched; R⁶ represents a C₁-C₁₀ alkyl group; and each of R⁷, R⁸, R⁹, R¹⁰, and R¹¹ represents a C₁-C₁₀ alkyl group, a C₁-C₁₀ alkenyl group, or a hydrogen atom independently to each other.

34. (New) A process for producing a hydrogenated ester according to claim 33, wherein the hydrogenating catalyst selected from at least one metal selected from the group consisting of Group VIII elements, Group IX elements, and Group X elements in the periodic table is at least one species selected from the group consisting of palladium, ruthenium and rhodium.